Inhomogeneous phase formation on the border of itinerant ferromagnetism

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A variety of analytical techniques suggest that quantum fluctuations lead to a fundamental instability of the Fermi liquid that drives ferromagnetic transitions first order at low temperatures. We present both analytical and numerical evidence that, driven by the same quantum fluctuations, this first order transition is pre-empted by the formation of an inhomogeneous magnetic phase. This occurs in a manner that is closely analogous to the formation of the inhomogeneous superconducting Fulde-Ferrel-Larkin-Ovchinnikov state. We derive these results from a field theoretical approach supplemented with numerical Quantum Monte Carlo simulations. Our analytical approach represents a considerable simplification over diagrammatic methods, makes contact with older analyses of the unitarity limit, and enables a simple physical picture to emerge.

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Many magnetic materials display second order ferromagnetic phase transitions. The transition temperature can be tuned using external parameters such as doping and pressure. Hertz realized that tuning such a transition to zero temperature could give rise to a new type of critical phenomena, for which he coined the term quantum criticality [1]. This led to a tremendous experimental and theoretical effort that has had some notable successes with the universal scaling predicted for the quantum critical regime being seen in a handful of materials [2]. However, in the majority of systems currently investigated, new behavior intervenes before the transition temperature can be tuned to zero. In many cases, such as $ZrZn_2$ [3], UGe_2 [4], MnSi [5], and CoS_2 [6], the second order transition becomes first order before the quantum critical point is reached. Moreover, recent experimental evidence points to phenomena that go beyond the first order transition; materials such as $ZrZn_2$ [3], UGe_2 [7], $Ca_3Ru_2O_7$ [8], $NbFe_2$ [9], and $Sr_3Ru_2O_7$ [10] all display unusual behavior in the vicinity of the putative quantum critical point.

This failure to find a naked quantum critical point has lead to speculation that it represents a fundamental principle [11]. Diagrammatic calculations that extend beyond the standard Moriya-Hertz-Millis [1] theory of itinerant quantum criticality suggest a breakdown of the Landau expansion around the quantum critical point [12, 13, 14]. This raises the question of how to connect the diagrammatic calculations to the well-established second order perturbation approach [15]. This approach accounts for all orders of vacuum scattering amplitude, and predicts that the itinerant ferromagnetic transition is first order at low temperature.

We show that, when a linearization of the electron dispersion about the Fermi surface is permissible, the first order magnetic transition is pre-empted by the formation of an inhomogeneous magnetic phase in a manner closely analogous to the inhomogeneous superconducting Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) state [16]. Our results are consistent with the effects of non-analyticities that appear in extensions to the Hertz-Millis theory [12, 13]. Here, we adopt an alternative field theoretical approach [17] to provide analytical evidence for our picture. As well as resolving the connection between the second order perturbation theory approach and the diagrammatic analysis, it provides insight into how quantum fluctuations drive the reconstruction of the phase diagram. Our analytical considerations are supported by Quantum Monte Carlo (QMC) simulations.

This letter is divided into two main parts: We first develop our analytical results and then discuss the QMC simulations that support them. Since one of the main advantages of our approach is the simplification that it affords, we outline the main steps of our analysis in their entirety. We follow this with a discussion of the effect of quantum fluctuations upon the phase diagram of the homogeneous ferromagnet – showing both how older results may be recovered from our analysis and the non-analyticities revealed by diagrammatics. Next, we show how the same fluctuations can drive a spatial modulation. After presenting our QMC results, we conclude with a discussion of possible extensions to the work.

In metals, the long-range component of the Coulomb interaction is screened. We therefore take as our starting point a free electron system interacting through a contact (Hubbard-like) repulsive interaction, g [1]. The corresponding partition function may be expressed as a fermionic coherent state path integral $\mathcal{Z} = \mathrm{Tr}\,\mathrm{e}^{-\beta(\hat{H}-\mu\hat{N})} = \int \mathcal{D}\psi\,\mathrm{e}^{-S}$ with the action

$$S = \int \sum_{\sigma=\pm} \bar{\psi}_\sigma \left(\partial_\tau + \hat{\zeta}\right) \psi_\sigma + \int g\bar{\psi}_+\bar{\psi}_-\psi_-\psi_+ \,. \quad (1)$$

Here $\int \equiv \int_0^\beta d\tau \int d^3x$, $\hat{\zeta}_{\hat{\mathbf{k}}} = \epsilon_{\hat{\mathbf{k}}} - \mu$, where $\epsilon_{\hat{\mathbf{k}}}$ denotes the dispersion and μ represents the chemical potential. To develop an effective Landau theory of the magnetic tran-

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sition, Hertz introduced a scalar Hubbard-Stratonovich decoupling of the interaction in the spin channel [1]. However, this form of decoupling neglects the potential impact of soft transverse field fluctuations. It is the latter that are responsible for driving the second order transition first order and in turn promote the instability towards inhomogeneous phase formation. Therefore, we will introduce a general Hubbard-Stratonovich decoupling which incorporates fluctuations in all of the spin $\mathbf{\Phi}$ and charge ρ sectors.

Defining $\mathbf{\Phi} = \mathbf{m} + \boldsymbol{\phi}$, where $\mathbf{m}$ denotes the putative saddle-point value of the magnetization field and $\boldsymbol{\phi}$ the fluctuations, one obtains the partition function $\mathcal{Z} = \int \mathcal{D}\psi\mathcal{D}\rho\mathcal{D}\boldsymbol{\phi}\mathrm{e}^{-S}$ where the action now takes the form $S = \int g(m^2+\phi^2-\rho^2)+\bar{\psi}[\partial_\tau+\hat{\zeta}+g\rho-g\boldsymbol{\sigma}\cdot(\mathbf{m}+\boldsymbol{\phi})]\psi$ [18]. We wish to study the potential for spatially modulated order where the magnetization forms a conical spiral, rotating about some axis (set along z) with a pitch vector $\mathbf{q}$. To simplify the analysis, it is helpful to transform to a rotating basis in which the magnetization becomes spatially uniform, $\mathbf{m} = (m_\perp, 0, m_\parallel)$. Setting $\psi \mapsto \mathrm{e}^{\mathrm{i}\mathbf{q}\cdot\mathbf{r}\,\sigma_z/2}\psi$,

$$\begin{pmatrix}\phi_\mathrm{x}\\ \phi_\mathrm{y}\\ \phi_\mathrm{z}\end{pmatrix} \mapsto \begin{pmatrix}\cos(\mathbf{q}\cdot\mathbf{r}) & -\sin(\mathbf{q}\cdot\mathbf{r}) & 0\\ \sin(\mathbf{q}\cdot\mathbf{r}) & \cos(\mathbf{q}\cdot\mathbf{r}) & 0\\ 0 & 0 & 1\end{pmatrix}\begin{pmatrix}\phi_\mathrm{x}\\ \phi_\mathrm{y}\\ \phi_\mathrm{z}\end{pmatrix},$$

and integrating over the fermion degrees of freedom yields the action

$$\begin{aligned} S = &\int g(m^2+\phi^2-\rho^2)\\ &-\mathrm{Tr}\ln[\underbrace{\partial_\tau+\hat{\zeta}_{\mathbf{k}+\sigma_z\mathbf{q}/2}-g\boldsymbol{\sigma}\cdot\mathbf{m}}_{\hat{G}^{-1}}+g\rho-g\boldsymbol{\sigma}\cdot\boldsymbol{\phi}]\,. \end{aligned} \quad (2)$$

Then, integrating over ρ and ϕ, an expansion of the action to second order in g leads to

$$\mathcal{Z} = \exp\left[-\int gm^2 + \mathrm{Tr}\ln\hat{G}^{-1} - \mathrm{Tr}\,g\hat{\Pi}^{+-} - \frac{1}{2}\mathrm{Tr}\,g^2(\hat{\Pi}^{+-}\hat{\Pi}^{-+}-\hat{\Pi}^{++}\hat{\Pi}^{--})\right],$$

where $\hat{\Pi}^{ss'} = \hat{G}^s\hat{G}^{s'}$ denotes the Lindhard function and $\hat{G}^{\pm} = (\partial_\tau + \epsilon^{\pm}_{\hat{\mathbf{k}},\mathbf{q}} - \mu)^{-1}$ with

$$\epsilon^{\pm}_{\mathbf{k},\mathbf{q}} = \frac{\epsilon_{\mathbf{k}+\mathbf{q}/2}+\epsilon_{\mathbf{k}-\mathbf{q}/2}}{2} \pm \frac{\sqrt{(\epsilon_{\mathbf{k}-\mathbf{q}/2}-\epsilon_{\mathbf{k}-\mathbf{q}/2}+2gm_\perp)^2+(2gm_\parallel)^2}}{2} \quad (3)$$

representing the energy of the electrons in plane-wave states with momentum $\mathbf{k}$ and spin-up or down relative to the mean-field spiral. To remove the unphysical ultraviolet divergence due to the contact nature of the interaction potential and arising from the term in the action second order in g, we must affect the standard regularization of the linear term $\mathrm{Tr}\,g\hat{\Pi}^{+-}$ setting $g \mapsto 2k_\mathrm{F}a/\pi\nu - 2(2k_\mathrm{F}a/\pi\nu)^2/V\sum'_{\mathbf{k}_{3,4}} n(\epsilon^+_{\mathbf{k}_3,\mathbf{q}})n(\epsilon^-_{\mathbf{k}_4,\mathbf{q}})(\epsilon^+_{\mathbf{k}_1,\mathbf{q}} + \epsilon^-_{\mathbf{k}_2,\mathbf{q}} - \epsilon^+_{\mathbf{k}_3,\mathbf{q}} - \epsilon^-_{\mathbf{k}_4,\mathbf{q}})^{-1}$ [19], where the prime indicates that the summation is subject to the momentum conservation $\mathbf{k}_1+\mathbf{k}_2 = \mathbf{k}_3+\mathbf{k}_4$. This regularization allows us to characterize the strength of the interaction through the dimensionless parameter $k_\mathrm{F}a$, where k_F denotes the Fermi wave vector, a is the s-wave scattering length, and ν is the density of states at the Fermi surface.

Finally, carrying out the Matsubara summations, one obtains the following expression for the free energy:

$$\begin{aligned} F = &\sum_{\mathbf{k},s}\epsilon^s_{\mathbf{k},\mathbf{q}}n\left(\epsilon^s_{\mathbf{k},\mathbf{q}}\right) + \frac{2k_\mathrm{F}a}{\pi\nu V}N^+_\mathbf{q}N^-_\mathbf{q}\\ &-2\left(\frac{2k_\mathrm{F}a}{\pi\nu V}\right)^2\sum_\mathbf{k}\int d\epsilon^+d\epsilon^-\frac{\rho^+_\mathbf{q}(\mathbf{k},\epsilon^+)\rho^-_\mathbf{q}(-\mathbf{k},\epsilon^-)}{\epsilon^++\epsilon^-}\\ &+2\left(\frac{2k_\mathrm{F}a}{\pi\nu V}\right)^2\sum_{\mathbf{k}_{1,2,3,4}}{}'\frac{n(\epsilon^+_{\mathbf{k}_1,\mathbf{q}})n(\epsilon^-_{\mathbf{k}_2,\mathbf{q}})}{\epsilon^+_{\mathbf{k}_1,\mathbf{q}}+\epsilon^-_{\mathbf{k}_2,\mathbf{q}}-\epsilon^+_{\mathbf{k}_3,\mathbf{q}}-\epsilon^-_{\mathbf{k}_4,\mathbf{q}}}\,, \end{aligned} \quad (4)$$

where $n(\epsilon) = 1/(1+\mathrm{e}^{\beta(\epsilon-\mu)})$ is the Fermi distribution, $N^s_\mathbf{q} = \sum_\mathbf{k} n(\epsilon^s_{\mathbf{k},\mathbf{q}})$, and $\rho^{\pm}_\mathbf{q}(\mathbf{k},\epsilon) = \sum_{\mathbf{k}'} n(\epsilon^{\pm}_{\mathbf{k}'+\mathbf{k}/2,\mathbf{q}})[1 - n(\epsilon^{\pm}_{\mathbf{k}'-\mathbf{k}/2,\mathbf{q}})]\delta(\epsilon - \epsilon^{\pm}_{\mathbf{k}'+\mathbf{k}/2,\mathbf{q}} + \epsilon^{\pm}_{\mathbf{k}'-\mathbf{k}/2,\mathbf{q}})$ is the spin-up(down) particle-hole density of states.

Spatially Uniform Case: To orient our discussion, we first consider the implications of the fluctuation corrections on the magnetic phase diagram for a free particle dispersion $\epsilon_\mathbf{k} = k^2/2m^*$ without accounting for spatial modulation. In this case Eq. (4) reduces to that obtained in Ref. [15] from second order perturbation theory. From this result, one finds that fluctuations drive the second order ferromagnetic transition first order at temperatures below that of the tricritical point, $T_\mathrm{T} \approx 0.2T_\mathrm{F}$, where T_F is the Fermi temperature (see Fig. 1). Substituting the low energy form of the particle-hole density of states, $\rho^{\pm}_{\mathbf{q}=\mathbf{0}}(\mathbf{k},\epsilon) = \epsilon\theta(kk^{\pm}_\mathrm{F} - k^2/2 - \epsilon)/2\pi k$, where $k^{\pm}_\mathrm{F} = \sqrt{2m^*(\mu \pm 2k_\mathrm{F}am/\pi\nu)}$, into the fluctuation correction to the free energy, and expanding in powers of magnetization, one recovers a singular term of order $m^4\ln m^2$ arising from particle-hole excitations with momentum near to $2k_\mathrm{F}$, the same non-analyticity as was found diagrammatically in Refs. [12, 13]. The formation of a finite magnetization increases the phase-space available for the formation of virtual intermediate pairs of particle-hole pairs, and this phase space enhancement ultimately drives the ferromagnetic transition first order.

Spatially Modulated Case: Quantum fluctuations also lead to spatial modulation of the magnetic order and further reconstruction of the magnetic phase diagram. The results of this analysis are shown in Fig. 1, which is obtained from Eq. (4) through a Landau expansion in m [20]. At low temperatures and $k_{\mathrm{F}}a = 0.84$, a second order transition into an inhomogeneous spin phase with pitch $0.1k_{\mathrm{F}}$ pre-empts the first order phase transition at $k_{\mathrm{F}}a = 1.055$. The expansion cannot however describe how far the textured phase penetrates into the uniform ferromagnetic phase. For a quadratic dispersion, the magnetization and spiral wave-vector enter the single electron energy in the combination $(\mathbf{k}\cdot\mathbf{q}/m^*)^2 + (4k_{\mathrm{F}}am_\perp/\pi\nu)^2$, where we have restricted to a planar spiral since it always has lower energy than a conical spiral at zero magnetic field. Upon linearizing the electron energy at the Fermi surface, q^2 and $m_\perp^2$ enter the free energy in the same way (up to the angular factors that accompany $\mathbf{q}$ and which are integrated over). Therefore, the spatial modulation enters as if it where a direction dependent magnetization. As a consequence, the coefficient of $m_\perp^4$ in our Taylor expansion is proportional to the coefficient of $q^2m_\perp^2$. When the $m_\perp^4$-term becomes negative – and a first order transition becomes favorable – the $q^2m_\perp^2$-term also becomes negative favoring a spatial modulation that emerges from the tricritical point. This is precisely the same situation as seen in the FFLO state [16].

To assess the validity of the perturbative scheme, we turn now to the numerical Quantum Monte Carlo analysis of the Stoner Hamiltonian (1) making use of the CASINO program [21]. These methods are based upon optimizing a trial wave function and are restricted to zero temperature. Our approach mirrors that used in previous studies of itinerant ferromagnetism [22, 23, 24]. The variational wave function used in our simulation, $\psi = D\mathrm{e}^{-J}$, is a product of a Slater determinant, D, that takes account of the Fermion statistics and occupation of single particle orbitals, and a Jastrow factor, J, that accounts for electron correlations.

The Slater determinant consists of plane-wave spinor orbitals containing both spin-up and spin-down electrons, $D = \det(\{\psi_{\mathbf{k}\in k_\uparrow}, \bar{\psi}_{\mathbf{k}\in k_\downarrow}\})$. Although not an eigenstate of total spin, the constituent spin states of least weight provide the dominant contribution to the variational state energy. In the case of uniform magnetization, for computational efficiency, we factorize the Slater determinant into an up and a down-spin determinant [21]. The spin textured phase is described by non-collinear spins, which have only recently been studied within the Variational Monte Carlo (VMC) [25]. These studies lead us to describe a planar spin spiral with a trial wave function that contains the spinors $\psi_{\mathbf{k}} = \mathrm{e}^{\mathrm{i}\mathbf{q}\cdot\mathbf{r}/2}(\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}}, \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{r}})$ and $\bar{\psi}_{\mathbf{k}} = \mathrm{e}^{-\mathrm{i}\mathbf{q}\cdot\mathbf{r}/2}(-\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}}, \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{r}})$ which explicitly fix the spin spiral orientation. For $\mathbf{q} = \mathbf{0}$ this would recover the factorized form for the Slater determinant employed in the uniform case. Our simulations are carried out in a unit cell with periodic boundary conditions commensurate with the pitch of the spiral.

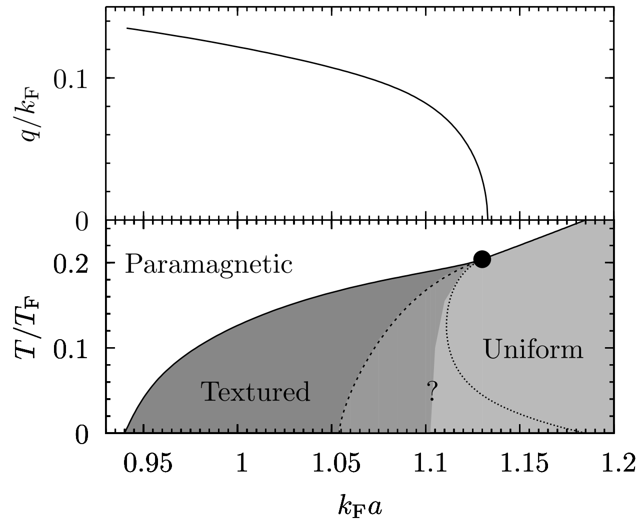

FIG. 1: The lower phase diagram shows the first order (dashed line) and second order (upper solid line) ferromagnetic ordering if the system were restricted to be homogeneous. The first order transition is pre-empted by a second order phase transition into a modulated ferromagnetic state bounded by the solid line. The dotted line shows where the uniform ferromagnetic phase would form, if the transition were restricted to be second order; this putative phase boundary not only comes after the first order transition and joins smoothly to the second order phase boundary at temperatures above the tricritical point, but also has negative slope at $T = 0$, consistent with Refs. [13]. The upper graph shows the wave vector q of the modulated phase boundary.

The Jastrow factor, J, accounts for electron-electron correlations. It consists of polynomial and plane-wave expansions in electron-electron separation, and a polynomial backflow function. In the spiral case, the Jastrow factor is restricted to be spin independent to maintain the spin spiral orientation and the wave function antisymmetry. The trial wave functions were optimized using QMC methods. In the uniform case, the optimization was performed in two steps using VMC and Diffusion Monte Carlo, whereas only VMC calculations were performed for the textured state. To model the repulsive contact potential between the electrons we employ the modified Pöschl-Teller interaction [26] which has smooth edges so that the QMC configurations can sample it faithfully [27].

Firstly, constraining the magnetization to be spatially uniform, an estimate of the ground state magnetization for different interaction strengths recovers the expected phase diagram (Fig. 2), revealing a first order phase transition into the itinerant magnetic phase. This provides a platform upon which to construct the full textured phase diagram. Allowing for spatial modulation of the magnetization, we find that an inhomogeneous magnetic phase pre-empts the transition into the uniform phase. The resulting textured phase has similar extent and wave vector to the analytical prediction lending support to the con-

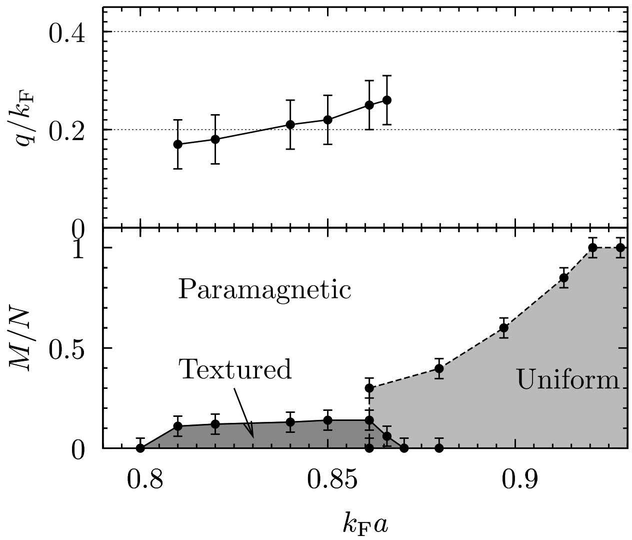

FIG. 2: The lower panel shows the variation of the ground state magnetization M with interaction strength $k_F a$ at zero temperature. The dashed line corresponds to the uniform phase, and the solid line is the textured phase. The upper panel shows the wave vector of the inhomogeneous magnetic phase, and the discrete values of q sampled in the investigation are highlighted by the horizontal dotted lines.

clusions of the perturbative field theoretic analysis.

In conclusion, quantum fluctuations are known to drive the itinerant ferromagnetic transition first order. We have shown that the same mechanisms lead to the development of inhomogeneous magnetic order in the vicinity of the tricritical point, resulting in a phase diagram which mirrors that of the superconducting FFLO phase. Our results are consistent with recent diagrammatic analyses and reveal the connection between these works [12, 13] and older second order perturbation calculations [15]. Moreover, our approach represents a considerable analytical simplification and more clearly reveals the underlying physical processes.

There are several directions in which this analysis might be extended. An electronic band dispersion can drive a similar reconstruction of the electronic phase diagram [28]. It would be informative to investigate the interplay between such effects and quantum fluctuations. As well as the possibility of spatially modulated magnetism, recent works have suggested that a band dispersion might lead to d-wave distortion of the Fermi surface [29]. Such electron nematics may be viewed as melted versions of the spatially modulated magnetism that we consider. Indeed, quantum fluctuations might drive the formation of an electron nematic phase in itinerant ferromagnets.

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